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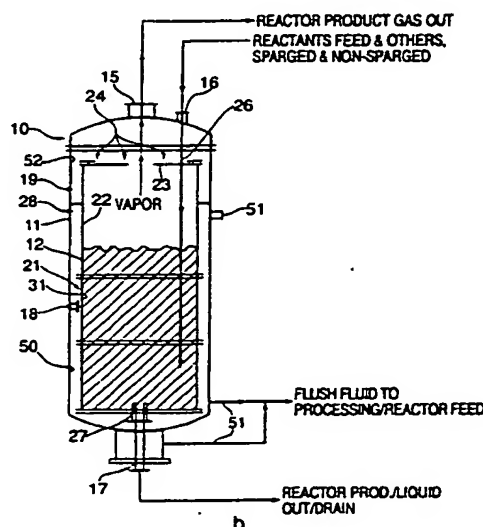
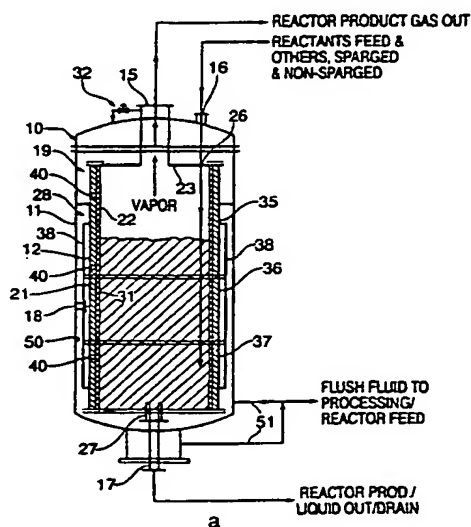
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(54) Title: APPARATUS FOR FLUORINATION OF ORGANIC COMPOUNDS

**(57) Abstract**

A reactor for fluorinating an organic compound comprising (a) an outer vessel (11); (b) a reactor vessel (12) being disposed within said outer vessel (11) to define an annular space (19), said reactor vessel (12) at least partially comprising a fluoropolymer, said annular space (19) being adapted to receive a fluid; (c) at least one pathway (51) for introducing said heating fluid into said annular space; (d) at least one pathway (26) for inputting reaction materials into said reactor vessel; and (e) at least one pathway for outputting (27) a product stream from said reactor vessel.

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APPARATUS FOR FLUORINATION OF ORGANIC COMPOUNDS

FIELD OF THE INVENTION

5 The invention relates generally to a system and method for reacting corrosive materials under pressure. More specifically, the invention relates to a reactor system and method for the liquid-phase fluorination of organic compounds.

BACKGROUND OF THE INVENTION

10 Liquid-phase fluorination involves a mixture of corrosive reaction materials. The corrosion is acute especially where Lewis-acid catalysts, such as antimony halide catalysts, are used under high reaction pressures and at elevated temperatures. Under these conditions, strong acids form which tend to corrode reactor vessels, even those comprised of corrosion-resistant materials. Reactor
15 corrosion compromises the structural integrity of the reactor and reduces its useful life. Therefore, a need exists to minimize reactor corrosion.

 A recent approach to combat reactor corrosion involves lining or coating the inside of the reaction vessel with a fluoropolymer. Although effective in preventing chemical corrosion, the fluoropolymer component of the reactor tends
20 to be problematic, especially for commercial-scale reactors, for example, over 500 gallons. Commonly encountered problems include body flange seal leakage, liner flexing stresses, hydrogen fluoride permeation through the fluoropolymer liner, and blister formation. Such problems diminish affect the reactor's durability and lead to premature failure. Additionally, the fluoropolymer component tends to
25 insulate the reactor thermally, necessitating costly and complex external heat transfer means and procedures.

 Thus, a need exists for non-corroding reactor systems that can be used for the commercial-scale production of fluorinated compounds where high pressures are encountered and heat transfer are required.

BRIEF DESCRIPTION OF THE DRAWINGS

Figs. 1a and 1b illustrate closed and opened reactor vessel embodiments, respectively, of the reactor system of the invention.

5 Fig. 2 illustrates an embodiment of the baffling means of the reactor system of the present invention.

Fig. 3 illustrates another embodiment of the baffling means of the reactor system of the present invention.

DETAILED DESCRIPTION OF THE INVENTION AND PREFERRED EMBODIMENTS

10 The present invention provides for a chemically-resistant reactor system that is durable and allows for efficient heat transfer. More specifically, the reactor system comprises a fluoropolymer-enhanced reactor vessel contained within a pressure-bearing outer vessel. The configuration of a vessel within another vessel defines an annular space. The combination of the pressure-bearing outer vessel
15 and the annular space facilitates heat transfer and enables the reactor vessel to vent and avoid blistering and other problems associated with fluoropolymer liners.

The reactor apparatus of the present invention facilitates heat transfer several ways. First, the annular space is adapted to contain one or more fluids, either liquid or gas, which may be externally heated or cooled and circulated
20 within the space. The annular space may also be baffled to impart velocity to the fluid to further enhance heat transfer.

In addition to accommodating a heat-transfer fluid, the annular space also allows for a thinner reactor vessel wall which tends to improve heat transfer. More specifically, since the outer vessel is constructed to be the pressure-bearing
25 component of the reactor system, the annular space can be pressured to approximately the reaction pressure. This results in near pressure equalization on either side of the reactor vessel wall. Consequently, the reactor vessel wall does not encounter forces from high pressure differentials and can be constructed with

relatively thin walls or it can be constructed totally of a fluoropolymer despite the fluoropolymer's marginal strength.

5 The annular space not only provides means of conveniently heating or cooling the reaction, but also improves the reactor vessel's useful life. In a preferred embodiment, the reactor comprises a rigid member and a loose, fluoropolymer liner. The rigid member has weep holes to allow those reactants that permeate the fluoropolymer-lined reactor to vent. This prevents reactants from building up between the liner and the reactor wall and forming blisters or otherwise leading to liner failure. Such a configuration therefore improves the reactor vessel's durability and extends its useful life.

10 The annular space accommodates a fluid which flushes away reaction materials escaping from the reactor vessel, and, when pressurized to approximate reaction pressure, the fluid relieves the stress on the fluoropolymer liner spanning the weep hole. Additionally, the fluid in the annular space is easily monitored to detect the existence of a leak. In a preferred embodiment, the fluid used to both impart heat and capture escaping reactants is a starting material which is fed into the reactor after circulating about the annular space. This way, the errant reactants are returned the reactor.

20 Therefore, in accordance with the present invention, a commercial-scale reactor may be designed more for corrosion resistance than for strength because the pressure-bearing outer vessel relaxes the structural demands on the reactor vessel. The present invention is especially useful for the liquid-phase fluorination of chlorinated hydrocarbons in which the reaction is very corrosive to conventional metal reactors. However, the system also may be useful for other processes, which may not be particularly corrosive, but would benefit from improved heat transfer and/or leak detection.

25 One aspect of the invention is a reactor. In a preferred embodiment, the reactor comprises: (a) an outer vessel; (b) a reactor vessel being disposed within said outer vessel to define an annular space, said reactor vessel at least partially comprising a fluoropolymer, said annular space being adapted to receive a fluid;

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(c) at least one pathway for introducing said fluid into said annular space; (d) at least one pathway for inputting reaction materials into said reactor vessel; and (e) at least one pathway for outputting a product stream from said reactor vessel.

Another aspect of the invention is a method for producing a compound using the reactor. The process comprises: (a) providing a reactor of the present invention; (b) feeding said reactor vessel with reaction material; (c) reacting said reaction materials to produce said compound by operating said reactor vessel at one or more reaction pressures and at one or more reaction temperatures; and (d) regulating said one or more reaction temperatures by introducing either a heated or cooled fluid into said annular space.

Referring now to Figs. 1a & 1b, preferred embodiments of the reactor apparatus 10 is shown. The reactor apparatus 10 comprises a reactor vessel 12 contained by an outer vessel 11. As shown, the outer vessel 11 includes an inlet 16 for the introduction of reaction materials, an outlet 15 for the removal of the product vapor stream, and an outlet 17 for the removal of the reaction liquid stream. Outer vessel 11 is of a size and shape suitable to enclose reactor vessel 12, which is supported within outer vessel 11 by supporting means 18. Reactor vessel 12 has a top 23 which has an inlet 26 through which reaction feed materials are introduced. Reactor outlet 27 permits withdrawal of a liquid stream from reactor vessel 12. Reactor outer surface 21 and vessel inner surface 50 define annular space 19. The annular space accommodates a fluid that can heat or cool the reactor vessel, and that can absorb and flush away reaction materials that leak from the reactor. The outer vessel 11 comprises an inlet/outlet 51 for introducing or removing fluid from the annular space.

The outer vessel 11 is designed as the pressure containing component of the system. It therefore should be structurally suited for expected operating pressures. The outer vessel is designed preferably to withstand pressures of at least about 50 psig, more preferably of at least about 100 psig, and still more preferably of at least about 160 psig. The construction requirements for such pressurized vessels is well known in the art.

The outer vessel 11 may be constructed of any conventional material used for liquid-phase fluorination reactors, such as corrosion resistant metals or fluoropolymers. Suitable metals include, without limitation, cold rolled steel, carbon steel, INCONEL™, HASTELLOY™, and the like. In the preferred embodiment, the vessel is made of carbon steel for economic reasons. Additionally, a portion of the vessel may be lined with a fluoropolymer as discussed below in regard to the reactor vessel 12.

Reactor vessel 12 is designed primarily to be non-corroding and commercially viable. It is not designed as a high-pressure containment for the reaction. Consequently, it can be comprised of a fluoropolymer which imparts excellent chemical resistance, although marginal strength. As used herein, the terms "fluorinated polymer" and "fluoropolymer" are used interchangeably and broadly refer to any polymer, copolymer or blend of polymers having a fluoride atom in at least one of the monomers. Preferred materials include, for example, polytetrafluoroethylene, poly(vinylidene fluoride), ethylene-tetrafluoroethylene polymer, ethylene-hexafluoropropylene polymer, tetrafluoroethylene-hexafluoropropylene polymer, perfluoroalkoxy polymer, any modified version of the above-mentioned polymers, and blends of two or more thereof. Polytetrafluoroethylene or its modified version is more preferred.

The reactor vessel 12 may be comprised totally of a fluoropolymer or its interior surface 22 may be lined or coated with a fluoropolymer. Where reactor vessel 12 is constructed entirely of fluoropolymer, the reactor wall preferably has a thickness of from about 25 to about 100 mm, although any thickness may be used within the scope of the invention. In the preferred embodiment, however, reactor vessel 12 is made of a rigid member 31 having its interior surface coated or lined with a fluoropolymer 22 (shown in Fig. 1a only). The rigid member 31 is typically constructed of a metal such as, for example, cold rolled steel, carbon steel, INCONEL™, HASTELLOY™, and the like. Preferably, the vessel is made of carbon steel for economic reasons since corrosion protection is provided by the fluoropolymer liner. With such a configuration, the metal wall thickness is

between about 3 to about 6 mm and the fluoropolymer thickness is between below about 1 to about 35 mm, preferably from about 1 to about 6 mm.

In a more preferred embodiment, the fluoropolymer component of the reactor vessel is a *loose* fluoropolymer liner. The term "loose fluoropolymer liner" broadly refers to a liner which covers at least portion of metallic part of the reactor and which is fitted from a film or sheet of a fluoropolymer material. Such a liner provides a reactor with a chemically-resistant barrier that is more durable than molded-type, fluoropolymer liners. More specifically, a loose liner tends to be less porous than a molded liner and, thus, tends to maintain a seal for a longer time. Preferably, the sheet has a thickness of no less than about 0.7 mm which is thicker generally than most molded liners.

Also in the more preferred embodiment, the reactor wall has at least one weep hole 40 (shown in Fig. 1a only), and preferably a plurality of weep holes 40. Weep holes allow reactants that permeate the liner to vent into the annular space. This way, pressure does not build up between the rigid member and the liner and cause blisters. The number and size of the weep holes may vary according to reactor size and other variables, although a weep hole having a diameter from about 1/8 to about 1/2 inch is generally preferred. A reactor having one or more weep holes requires that the liner have adequate strength to span the weep hole under reaction pressures. However, since the annular space can be pressurized to near reaction pressure, the resultant pressure on the fluoropolymer liner is very little.

Reactor top 23 may be closed or opened, as shown in Figs 1a & 1b respectively. As shown in Fig. 1a, the reactor vessel has a closed top such that the contents of reactor vessel 12 do not enter annular space 19. In closed reactor vessel embodiments, a pressure balancing device 32 is preferable. The pressure balancing device 32 would typically be a pressure differential control valve.

Pressure balancing is desirable for two reasons. First, if a significant difference develops between the pressures of the reactor and the annular space, then the reactor needs to be constructed to withstand that differential. This results

in an increase in the reactor wall thickness. For example, to withstand full operating pressure of 160 psig, the reactor wall must be several times thicker than the preferred thickness of 3 to 6 mm. A thicker reactor wall, of course, decreases the heat transfer efficiency. Second, if the annular space pressure were higher than the reactor's, then there would be no force driving the reactant through the weep holes of the reactor and into the annular space. On the contrary, the fluid may even enter the weep holes and cause the liner to blister thereby magnifying the problems weep holes are designed to prevent.

It is therefore preferable to maintain a *slight* positive pressure in the reactor relative to the annular space. The pressure in the annular space should be about 0.1 to about 25 psig lower than the reactor. The pressure in the reactor vessel can be a negative or positive pressure relative to atmospheric, but preferably positive.

An opened reactor vessel 12, as shown in Fig. 1b, contains one or more outlets 24 permitting the flow of gaseous reaction materials, products and byproducts from reactor vessel 12 into annular space 19. Since the gaseous product stream tends to contain only small amounts of Lewis acid catalyst, it is not particularly corrosive. Nevertheless, in the event the liquid reactants overflow the reactor or just for extra corrosion protection, it is preferable to coat or line the upper space 52 of the inner surface 50 of the outer vessel 11 with a fluoropolymer.

Reactor vessel 12 may be of any desired size, however, to be commercially viable, it should be at least about 500 gallons, and preferably at least 1000 gallons. Such capacities are possible using a loose, fluoropolymer liner, which requires no special equipment for installation, such as a large rotary oven. Moreover, the durability of such large reactors is improved by the weep holes which prevent blisters from forming.

Reactor vessel 12 may be constructed by any convenient means known in the art. It may be preferable for reactor vessels larger than about 1000 gallons to be segmented. For example, reactor vessel 12 may be constructed of top 35,

middle 36, and bottom 37 segments which are connected together with any suitable connecting means such as by bolting flanges of each segment together.

Furthermore, heat transfer may be further increased by providing reactor vessel 12 with fins 38 or rods which increase the surface area available for heat transfer. For example, the fins 38 or rods may be welded or attached to the outside surface of the reactor vessel such that they extend into the annular space. Such fins or rods may even be configured to act as baffles as described below.

The disposition of the reactor vessel within the outer vessel defines an annular space. The annular space facilitates heating the reactor, contains reaction materials leaking from the reactor vessel, and substantially equalizes reactor pressure.

Annular space 19 contains fluid 28 that may be heated to supply heat to reactor vessel 12. Fluid 28 may be heated or cooled by any conventional means, such as a heat exchanger, and then circulated into annular space 19. Preferably, in order to increase the rate of heat transfer between fluid 28 and reactor vessel 12, a means for increasing the velocity of fluid 28 in annular space 19 is provided. Suitable means include, without limitation, baffles, paddles and circulating pumps.

Figs. 2 and 3 depict alternative velocity increasing means. Fig. 2 depicts a spiral baffle 41, and Fig. 3 depicts cascade baffles 42. As mentioned above, the baffles also increase available surface area to improve heat transfer between the reactor vessel and the fluid. To construct such baffles, one edge of the baffle may be welded onto the outside surface of the reactor vessel. Once the reactor vessel is inserted into the outer vessel, the other baffle edge will loosely fit against the inner wall of the outer vessel. Other construction techniques will be apparent to those skilled in the art.

The annular space also is useful for flushing away leaks from the reactor vessel. In a preferred embodiment, the reactor apparatus is designed to accommodate leaks of reaction materials, such as hydrogen fluoride, through the reactor fluoropolymer. More specifically, a free-standing fluoropolymer film is provided along reactor inner surface 22 and weep holes 40 are provided in reactor

5 wall. Any reactant leaking through the fluoropolymer will be absorbed or otherwise flushed away by fluid 28 in annular space 19. In a more preferred embodiment, a hydrogen fluoride sensor is provided to detect the presence and concentration of one or more reaction materials, such as hydrogen fluoride, in annular space 19. Any suitable sensor may be used such as an optical sensor.

10 The type of fluid used in the annular space depends upon the user's preference and heating/cooling requirements. Suitable fluids range from relatively inert substances, such as oils and glycols, to starting materials for the reaction, such as organic compounds. In the preferred embodiment, the fluid is a starting material which is subsequently fed into the reactor. This way, the fluid imparts heat to the reactor externally when circulating and internally when fed to the reaction. Moreover, any reactants which leak through the reactor that are absorbed by the fluid are returned to the reactor when the fluid is fed to the reaction.

15 The size of the annular space 19 also depends upon the preference of the user. A narrower annular space provides for better heat transfer, but is more difficult to construct. Therefore, in practice, it is a compromise between heat transfer efficiency and construction complexity. A typical width ranges from about 50 to about 200 mm, preferably from about 100 to about 150 mm.

20 The apparatus of the invention is well suited for reactions in which corrosive starting materials, catalysts, by-products, or products are involved. Moreover, the apparatus is particularly well suited where high temperature and pressures are also required. One such reaction is the liquid-phase fluorination of an organic compound to produce a fluorinated product. In such a process, the reaction materials, which include an organic starting material, a fluorinating agent, and a fluorinating catalyst, are charged to reactor vessel 12 via inlets 16 and 26. Alternatively, as described above, the organic starting material may first be circulated in the annular space to impart heat to reactor and absorb leaks, and then fed into the reactor to exploit its remaining heat, to return errant reaction materials, and to feed the reaction.

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The organic starting material may be any compound containing a carbon-bonded chlorine or other atom replaceable by fluorine and/or that may contain a carbon-carbon unsaturated bond that is saturatable with fluorine.

Illustrative organic starting materials include, without limitation, chlorinated hydrocarbon compounds containing from 1 to 6 carbon atoms and 1 to 12 chlorine atoms. Preferred organic starting material compounds have the formula $CF_yCl_{3-y}CH_2CHF_wCl_{2-w}$, wherein $w=0$ or 1 , and $y=0-3$ (see also, U.S. Patent No. 5,574,192).

Suitable fluorination agents include any material which provides fluorine for the fluorination reaction. Preferred fluorination agents include, for example, hydrogen fluoride, elemental fluorine, BF_3 , antimony pentafluoride. The more preferred fluorination agent is hydrogen fluoride.

Any suitable fluorination catalyst may be used including, without limitation antimony, arsenic, niobium, tin, titanium, and tantalum halide catalysts. Examples of these catalysts include, without limitation: pentavalent antimony, niobium, arsenic and tantalum halides; pentavalent antimony, niobium, arsenic and tantalum mixed halides; and mixtures of pentavalent antimony, niobium, arsenic and tantalum halide catalysts. Pentavalent antimony, niobium, arsenic and tantalum halides are commercially available, and mixed halides thereof are created *in situ* upon reaction with hydrogen fluoride (see Patent No. 5,574,192). Antimony pentachloride is more preferred due to its low cost and availability. Pentavalent antimony mixed halides of the formula $SbCl_nF_{5-n}$ and $SbBi_nF_{5-n}$ where n is 0 to 5 are most preferred. Although the amount of fluorination catalyst used may vary widely, the weight percent of catalyst relative to the organic starting material generally ranges from about less than 1 to about 75%, preferably from about 5 to about 50%, and more preferably from about 10 to about 25%.

It may be advantageous to periodically regenerate the catalyst which may be accomplished by any means well known in the art. For example, the catalyst may be regenerated by adding chlorine, in an amount from about 1 to about 10 mole percent relative to the amount of catalyst initially present in the reactor, to the combination stream of organic starting material and the recycled stream of

under-fluorinated materials and hydrogen fluoride. The chlorine may be continuously or intermittently added. One of ordinary skill in the art can readily determine the amount of chlorine to be added in order to optimize the use of the catalyst.

5 The organic starting material is preheated, preferably outside the reactor to a temperature of from about ambient to about 350°F. The organic starting material may be used as fluid 28 which is circulated in annular space 19, in which case the organic material is reheated to a temperature just below the temperature at which the compound breaks down so as to impart the maximum heat via heat
10 exchange to the reaction mixture. For example, in the case of using G240fa as the organic starting material, the preheat temperature is likely to be limited to about 250°F to minimize breakdown. Absent thermal breakdown, higher temperatures are preferred up to about 350°F which is just below the thermal property limits of many fluoropolymer components.

15 Reaction pressure can vary and optimal pressures can be determined by someone skilled in the art without undue experimentation. Convenient operating pressure range from about 30 to about 300 psig, preferably from about 70 to about 260 psig, and more preferably from about 100 to about 200 psig. As mentioned
20 above, the pressure in the annular space preferably should be less, and preferably about 0.1 to about 25 psig below the reaction pressure to maintain the reactor at a slight positive pressure.

 The reaction temperature generally is from about ambient to about 350°F, preferably from about 100 to 200° F. Heat can be supplied either by introducing superheated HF at about 200 to about 400° F, or by circulating fluid 28 at least
25 partially in the annular space at an initial temperature of about 200 to about 400° F, and preferably about 250 to 350°F.

 Reaction times are dependent on several factors including catalyst concentration, the type of catalyst, and the temperature. For a batch process, the progress of the reaction can be monitored conveniently by the increase in pressure
30 due to the formation of by-product HCl. Typical reaction times range from about

1 to about 25 hours, and preferably from about 2 to about 8 hours. For a continuous process, the reaction times ranges from about 1 second to about 5 hours, and, preferably, from about 10 seconds to about 1 hour.

5 The fluorination reaction provides a product stream from which the fluorinated product is recovered. In a preferred embodiment, the fluorinated product is recovered by distillation techniques well known in the art.

The invention will be clarified further by a consideration of the following non-limiting example.

10 EXAMPLE 1

This example illustrates the production of 1,1,1,3,3-pentafluoropropane (245fa), which is highly corrosive, in a 2000 gallon reactor vessel, which is generally considered viable for commercial-scale production.

15 The reaction apparatus used comprised a 2000 gallon steel reactor vessel lined with a loose, 3 mm thick PTFE sheet, and a 3000 gallon outer vessel which supported and enclosed the reactor vessel. To the reactor vessel were introduced: $C_3H_3Cl_3$ in the liquid phase, at a temperature of 200°F, at pressure of 160 psig, and at rate of about 4500 lb/hr; and HF at a temperature of 350°F, at pressure of 160 psig, and at rate of 3300 lb/hr. The reactor vessel was pre-charged
20 with 10,000 lb of antimony pentachloride catalyst.

The product, 245fa (2700 lb/hr), along with HCl (3500 lb/hr) and unreacted starting material (about 1500 lb/hr) were removed in a gaseous product stream and separated using conventional distillation techniques.

CLAIMS

WHAT IS CLAIMED IS:

1. A reactor for fluorinating an organic compound comprising:
an outer vessel;
5 a reactor vessel being disposed within said outer vessel to define an annular space, said reactor vessel at least partially comprising a fluoropolymer, said annular space being adapted to receive a fluid;
at least one pathway for introducing said fluid into said annular space;
at least one pathway for inputting reaction materials into said reactor vessel;
10 and
at least one pathway for outputting a product stream from said reactor vessel.
2. The reactor of claim 1, wherein said reactor vessel is configured to
15 accommodate an internal pressure less than said outer vessel.
3. The reactor of claim 2, wherein said outer vessel is configured to accommodate an internal pressure of at least about 50 psig.
- 20 4. The reactor of claim 1, wherein said reactor vessel comprises a rigid member having an interior surface at least partially lined with said fluoropolymer, said rigid member having at least one weep hole in fluid communication with said annular space

5. The reactor of claim 4, wherein said fluoropolymer liner has a thickness of from below about 1 to about 6 mm and wherein said rigid member has a thickness of about 3 to about 6mm.
- 5 6. The reactor of claim 1, wherein said apparatus also comprises baffles in said annular space.
7. The reactor of claim 1, wherein said annular space and the interior space of said reactor vessel are in fluid communication.
- 10 8. The reactor of claim 7, wherein said reactor vessel is open to said annular space.
- 15 9. The reactor of claim 8, wherein said outer vessel is at least partially lined with a fluoropolymer.
10. The reactor of claim 7, wherein said reactor vessel is closed to said annular space and comprises a regulator valve to regulate the pressure differential between said interior space and said annular space.
- 20 11. The reactor of claim 1, wherein said reactor vessel has a capacity of at least 500 gallons.
- 25 12. The reactor of claim 1, wherein said reactor vessel has heat transfer members extending into said interior space.

13. The reactor of claim 1, further comprising:

leak detection means for monitoring said fluid to detect the presence of one or more reaction materials.

5 14. A method for producing a compound, said process comprising:
providing a reactor comprising at least:

an outer vessel; and

10 an reactor vessel defining an interior space and being
disposed within said outer vessel to define an annular
space between said outer vessel and said reactor
vessel, said annular space being adapted to receive a
fluid;

feeding said reactor vessel with reaction material;

15 reacting said reaction materials to produce said compound by operating said
reactor vessel at one or more reaction pressures and at one or more
reaction temperatures; and

regulating said one or more reaction temperatures by introducing either a
heated or cooled fluid into said annular space.

20 15. The method of claim 14, further comprising:

maintaining a reaction pressure no less than the pressure of said annular
space.

25 16. The method of claim 15, wherein said interior space is at a pressure from
below 1 to about 50 psig higher than the pressure of said annular space.

17. The method of claim 14, wherein said fluid comprises one or more reaction materials which are circulated at least partially in said annular space before being fed to said reactor vessel.

5 18. The method of claim 14, wherein an organic starting material is reacted with a fluorination agent in the presence of a fluorination catalyst.

19. The method of claim 18, wherein said interior space is at a pressure no less than about 100 psig.

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20. The method of 18, wherein said fluid is heated and comprises at least partially said organic compound and wherein said method further comprises:

feeding at least a portion of said heated fluid into said reactor vessel after circulating said heated fluid at least partially through said annular space.

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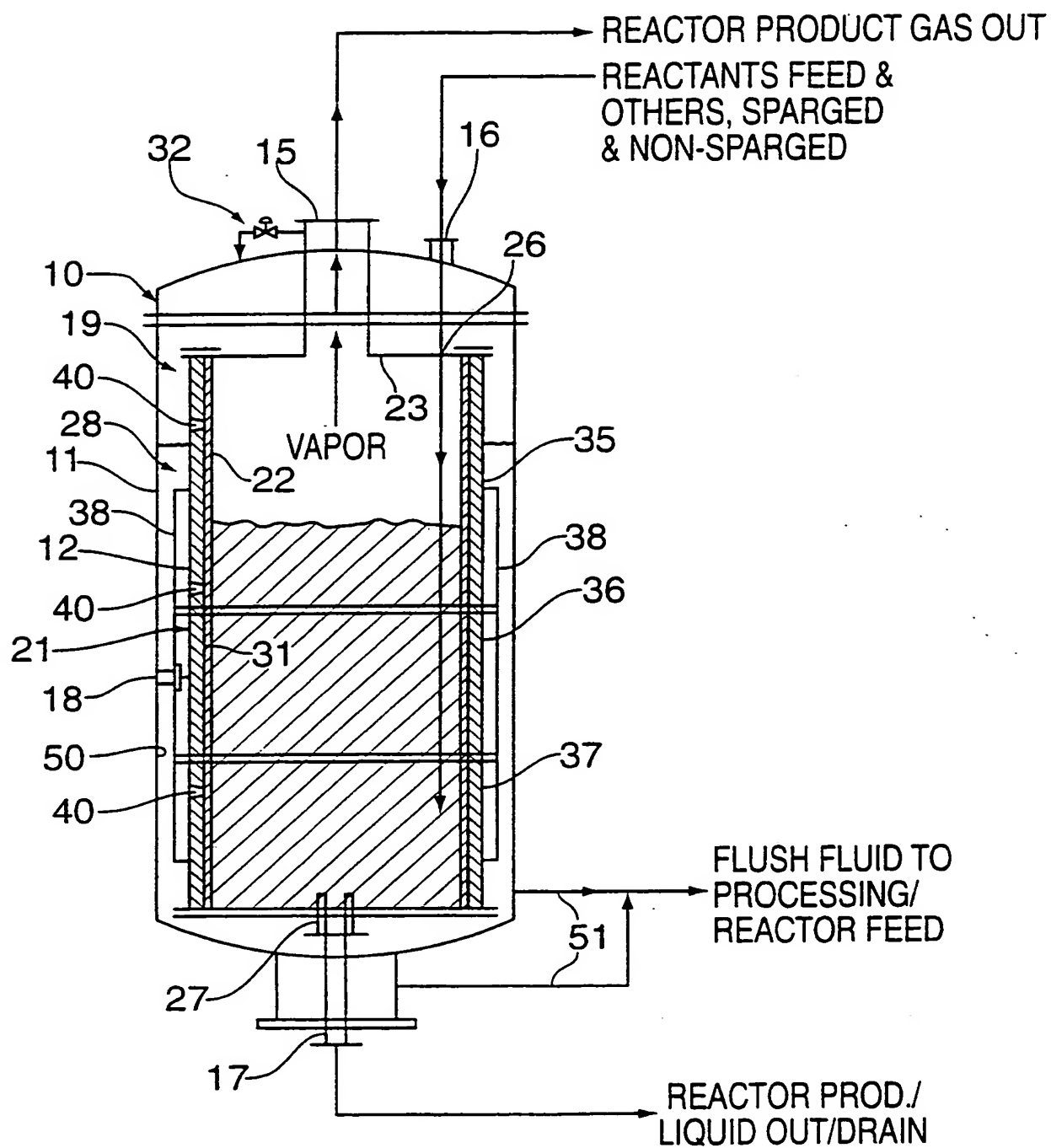


FIG. 1a

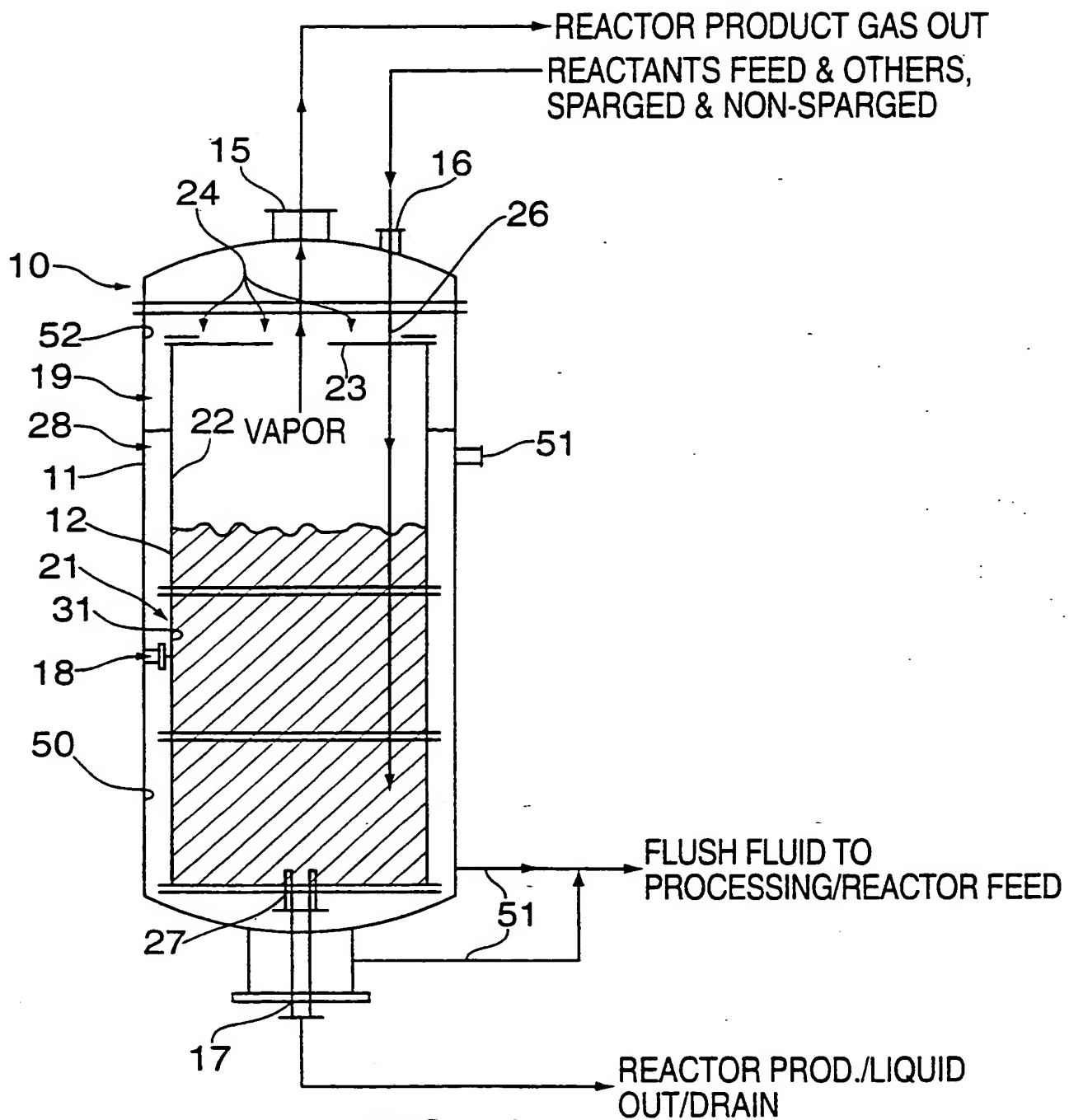


FIG. 1b

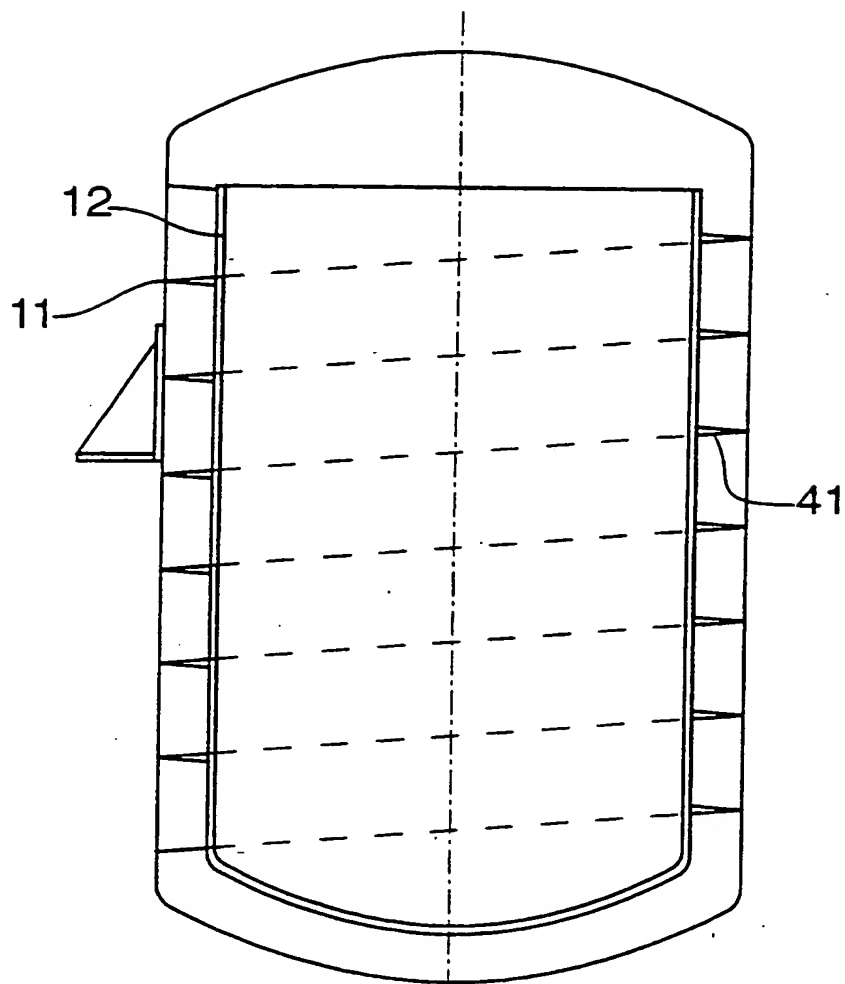


FIG. 2

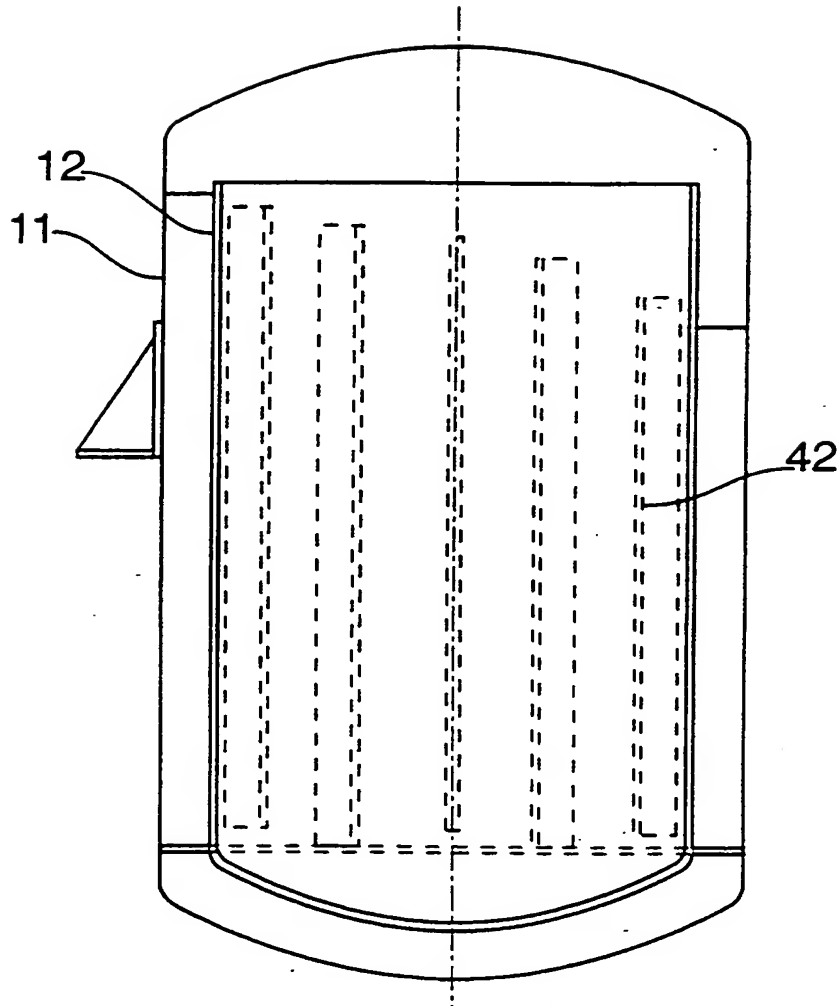


FIG. 3

INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 98/25031

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 B01J3/04 B01J19/02 B01J19/00 C07C17/20 C07C19/08

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 B01J C07C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE 43 11 417 A (WALTER LUDWIG BEHAELTER- UND STAHLBAU) 13 October 1994	14
Y	see column 3, line 10 - column 4, line 65	1,13
A	see figure 1	3

X	EP 0 099 443 A (UHDE GMBH) 1 February 1984	14
Y	see the whole document	15-18

Y	US 2 768 200 A (BUSBY LYLE E) 23 October 1956	15-17
A	see column 1, line 15 - column 3, line 14 see figure 1	14

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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
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Date of the actual completion of the international search

15 April 1999

Date of mailing of the international search report

26/04/1999

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Vlassis, M

INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 98/25031

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	CHEMICAL ABSTRACTS, vol. 123, no. 25, 18 December 1995 Columbus, Ohio, US; abstract no. 339132, KIMURA M, YOKOYAMA T: "Liquid-phase fluorination of organic compounds in fluoropolymer reactors" XP002077106 & JP07233102 A (ASAHI GLASS CO LTD; JAPAN) 5 September 1995 see abstract	1,13,18
A	US 3 472 632 A (HERVERT GEORGE L, WEBER HAROLD C) 14 October 1969 see the whole document	1,4
X	DE 388 127 C (ERDÖL- U. KOHLE-VERWERTUNG AKT.-GES) 10 January 1924	14
A	see page 3, left-hand column, line 34 - right-hand column, line 75 see figure 2	1,2

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 98/25031

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
DE 4311417	A	13-10-1994	NONE	
EP 0099443	A	01-02-1984	DE 3227545 A JP 59032932 A	26-01-1984 22-02-1984
US 2768200	A	23-10-1956	NONE	
US 3472632	A	14-10-1969	NONE	
DE 388127	C		NONE	

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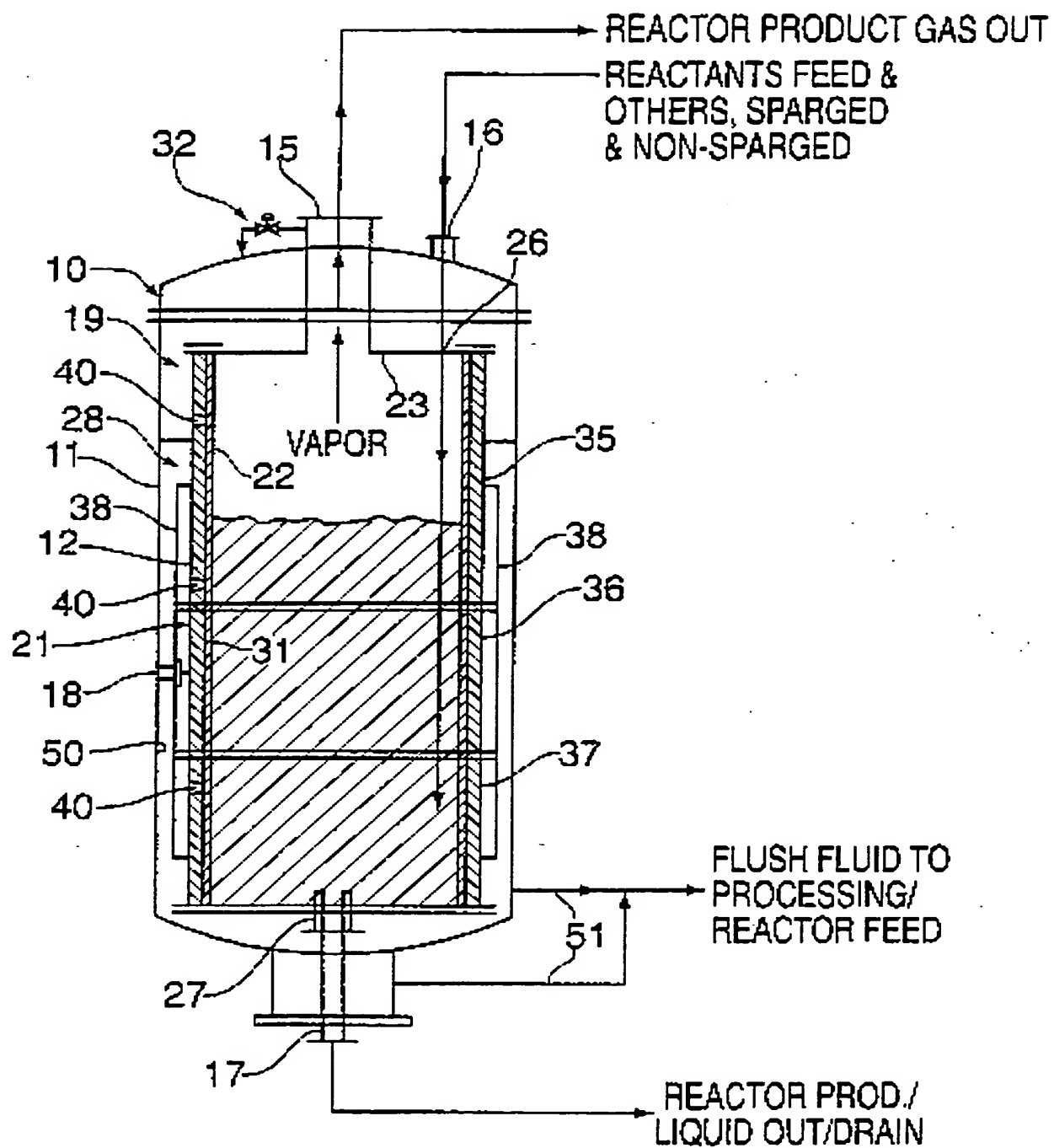


FIG. 1a

FIG. 1b

SUBSTITUTE SHEET (RULE 28)

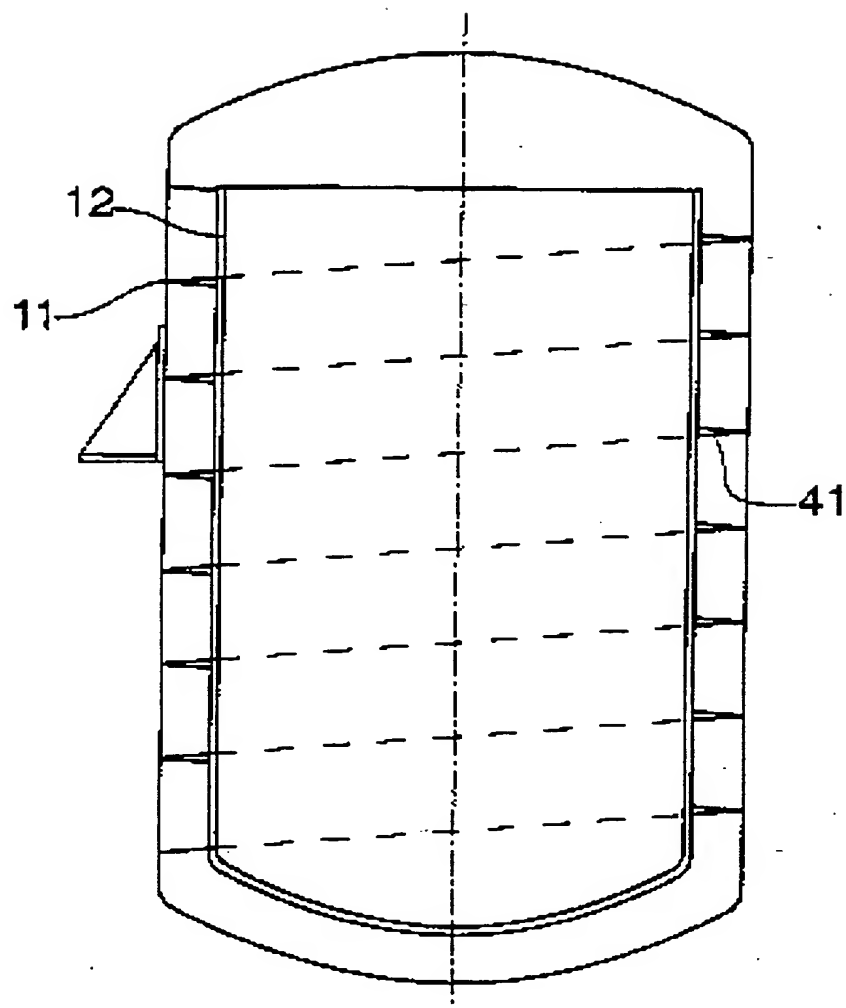


FIG. 2

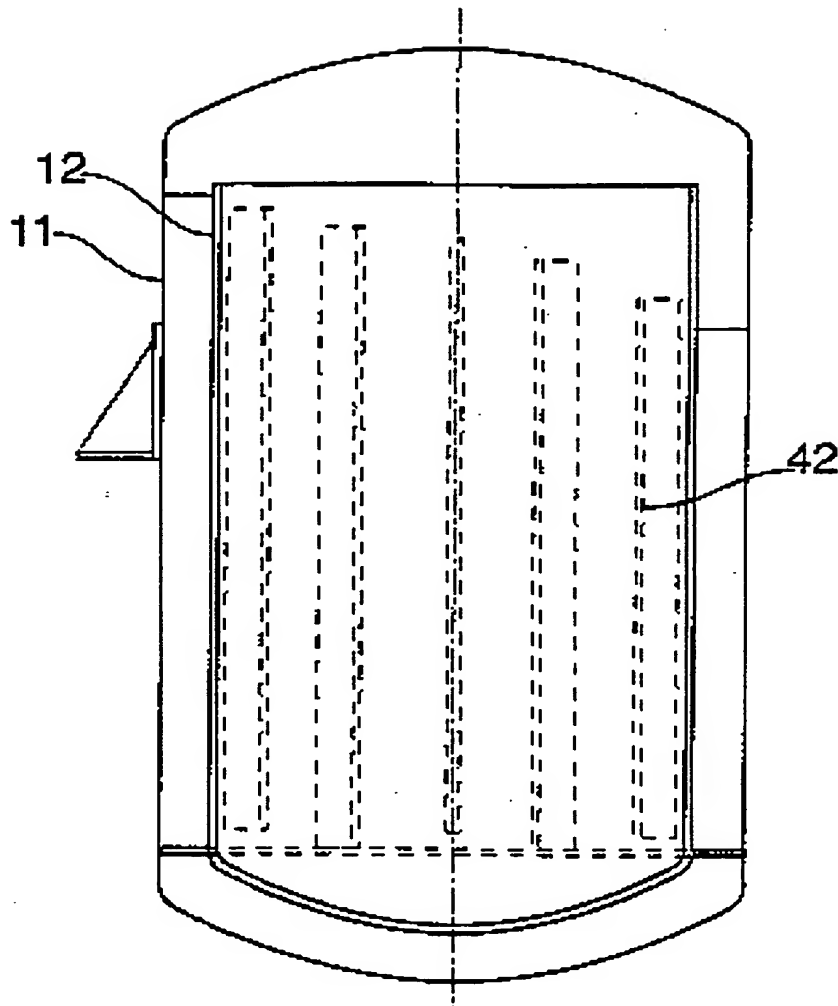


FIG. 3